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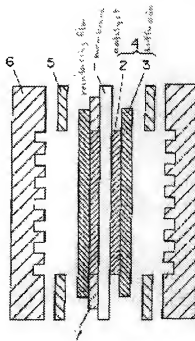
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(54) SOLID HIGH POLYMER TYPE FUEL CELL

(57)Abstract:

PROBLEM TO BE SOLVED: To prevent damage of an ion exchange membrane, and improve sealing performance of a fuel cell by setting a thickness of the ion exchange membrane to a specific thickness, making the area of a catalyst layer smaller than a diffusive layer in a gas diffusive electrode, making a peripheral part of the catalyst layer exist on the inside of the diffusive layer, and providing an architrave-shaped protective film at least on one surface side in the ion exchange membrane.

SOLUTION: The area of a catalyst layer 2 is made smaller than the area of a diffusive layer 3. A fuel cell comprises an ion exchange membrane 1 having a gas diffusive electrode 4 where a peripheral edge part of the catalyst layer 2 exists on the inside of a peripheral edge part of the diffusive layer 3 and an



architrave-shaped reinforcing film 7 where a window frame coincident with a shape of the catalyst layer 2 is arranged, at least one both sides of one. By such a constitution, an edge part of the electrode diffusive layer 3 can be made to coincide with a part where the reinforcing film 7 exists without reducing the contact area of the reinforcing film 7 and the ion exchange membrane 1 whose thickness is not more than 50 micron.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates especially to the ion exchange membrane of a polymer electrolyte fuel cell about a polymer electrolyte fuel cell.

[0002]

[Description of the Prior Art] The polymer electrolyte fuel cell (it is henceforth described as PEFC) uses the precious metal catalyst for the electrolyte for the ion exchange membrane which is a solid-state polyelectrolyte as an electrode catalyst.

[0003] When hydrogen is used as a fuel in PEFC, the reaction of (** 1) occurs in a negative electrode.

[0004]

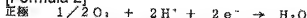
[Formula 1]



[0005] Moreover, when oxygen is used as an oxidizer, in a positive electrode, the reaction of (** 2) occurs and water is generated.

[0006]

[Formula 2]



[0007] If ion exchange membrane is not a certain water content more than fixed, and (** 1) (** 2) will not show ion conductivity, and a reaction will not occur. Moreover, if water content is low even if ion conductivity is shown, resistance of ion exchange membrane will become large and an output will decrease by IR loss. Therefore, in PEFC, in order to fully carry out the water of the ion exchange membrane, fuel gas and oxidant gas are humidified and supplied. In order that the water used for humidification may prevent contamination of the film or a catalyst, impurities, such as the organic substance, and a chlorine ion, a metal ion, must not be contained, but conductivity must be the pure water of 10 to 2 or less Scms. Since humidification water is consumed with actuation of PEFC, supply is needed. In order to exclude supply of this pure water, there is a system (United States patent No. 5,200,278) which is made to circulate through reactant gas and collects water, but in order to prevent mixing of an impurity, a system, such as preparing the filter of ion exchange resin, becomes complicated.

[0008] To application to the small noncommercial use power source of PEFC, in order to consider as a simpler system, humidify[no]-izing of reactant gas is desirable. However, ion exchange membrane is not fully humidified only by no humidifying-izing [reactant gas], but resistance becomes large, and a property falls. In order to solve this problem, the ion exchange membrane is made to humidify with the water which was made to carry out the

direct reaction of the hydrogen and oxygen which are made to contain noble metals and a metal oxidization particle, and cross in an ion exchange membrane, and generated them in the ion exchange membrane (JP,7-90111,A). At this time, 30-200 micrometers of thickness of ion exchange membrane are preferably set to 50-100 micrometers.

[0009] However, if the thickness of ion exchange membrane becomes thin at about 50 micrometers, the crossover to which reactant gas passes through ion exchange membrane will start, and an electrical potential difference will fall. Drawing 3 is the sectional view of the conventional polymer electrolyte fuel cell. The electrode 4 which becomes both sides of ion exchange membrane 1 from a catalyst bed 2 and the diffusion tub 3 is arranged, a gasket 5 is arranged at the periphery section of an electrode 4, a separator 6 **** these, and the cell is constituted. There is nothing that supports ion exchange membrane 1 between an electrode 4 and a gasket 5. Therefore, when the thickness of ion exchange membrane was thin, the following faults arose. Mechanical stress might start ion exchange membrane on the occasion of the assembly of a cell, and ion exchange membrane might be damaged. Moreover, at the time of actuation, expansion and contraction of the ion exchange membrane of the perimeter part of an electrode start by change and desiccation of the water content of a film-electrode zygote. Stress might be started and damaged to ion exchange membrane by this dimensional change. Furthermore, ion exchange membrane might be torn by the differential pressure of fuel gas and oxidant gas. Moreover, when joining an ion exchange membrane to an electrode with a hotpress, the ion exchange membrane might be damaged with the edge of the diffusion layer of an electrode. If ion exchange membrane is damaged as mentioned above, oxidant gas will be mixed with fuel gas and the serious problem for safety, such as burning on a catalyst, will arise.

[0010] Then, the following amelioration has been made in order to prevent membranous breakage. The periphery section of an electrode and the rim section of the ion exchange membrane by which an electrode is not arranged are covered. And arrange the reinforcement film of gas impermeability so that it may lap with a gas-seal part (JP,5-242897,A). Make it unify with a sealant using the sealant of the shape of liquefied or a sheet so that it may lap with the perimeter of a front face of the electrode of a film-electrode zygote (JP,8-45517,A). Join a plate-like reinforcement member to a film-electrode zygote using a sealant (JP,7-65847,A). one field side of ion exchange membrane is equipped with the protective coat of the sheet of the shape of a frame which is stuck by the periphery section of ion exchange membrane, and has a lap in an electrode at least (JP,5-21077,A --) Use the ion exchange membrane which put 5 No. -174845 official report and ion exchange membrane with small ion exchange capacity (it is described as henceforth E.W.) by the large ion exchange membrane of E.W., and was unified (JP,6-251780,A). It is the approach (JP,7-220742,A) of forming an auxiliary gasket in the periphery section of a diffusion layer by the sealant.

[0011]

[Problem(s) to be Solved by the Invention] However, when the thin ion exchange membrane of thickness was used, in order to reinforce with the configuration of above-mentioned JP,5-242897,A, JP,8-45517,A, and JP,7-65847,A at a film-electrode zygote, with it, it had the fault that breakage of the film with the edge of the diffusion layer at the time of joining the film to an electrode could not be prevented. Furthermore, with the configuration of JP,5-242897,A and JP,8-45517,A, since a protective coat or the seal section lapped with an electrode, it had the fault that the area to which reactant gas is supplied became smaller than an electrode surface product. Moreover, with the configuration of JP,5-21077,A and JP,5-174845,A which are shown in drawing 4, since an electrode 4 is joined to the ion exchange membrane 1 which has the reinforcement film 7, breakage of the ion exchange membrane 1 at the time of junction can be prevented. However, since the reinforcement film 7 existed between the electrode catalyst bed 2 and ion exchange membrane 1, it had the fault that the effective area of an electrode 4

and ion exchange membrane 1 decreased. Moreover, there is futility of joining expensive ion exchange membrane to a part without the ion conductivity need with the configuration of JP,6-251780,A, and the spreading process of a sealant is complicated with the configuration of JP,7-220742,A.

[0012] Moreover, with the above-mentioned conventional configuration for non-humidified operation, hydrogen was consumed by the chemical reaction and it had the fault that the utilization factor of hydrogen fell.

[0013] This invention solves such a conventional technical problem, also when a thin film is used, breakage of ion exchange membrane is prevented, and it aims at offering the polymer electrolyte fuel cell which raised seal nature. The polymer electrolyte fuel cell which operates by no humidifying is offered without furthermore reducing a hydrogen utilization factor.

[0014]

[Means for Solving the Problem] In order to solve the above-mentioned technical problem, as for the polymer electrolyte fuel cell of this invention, the thickness of ion exchange membrane uses a thing 50 micrometers or less, in a gas diffusion electrode, the area of a catalyst bed shall be smaller than the area of a diffusion layer, and there shall be the periphery section of a catalyst bed inside the periphery section of a diffusion layer, and ion exchange membrane shall have the protective coat of the shape of a frame which opened the aperture which is in agreement with the configuration of a catalyst bed in one [at least] field side.

[0015] Even when thickness uses the ion exchange membrane of 50 micrometers or less and a thin film by the above-mentioned configuration, breakage of ion exchange membrane can be prevented, without decreasing the effective area of an electrode and ion exchange membrane, and breakage of the film at the time of the assembly of a cell and actuation can be prevented, and seal nature can be raised.

[0016] Furthermore, the polymer electrolyte fuel cell of this invention is supplied to a positive electrode by oxidant gas, and supplies fuel gas to a negative electrode by no humidifying, respectively.

[0017] Since the absolute magnitude of the water for the concentration gradient of the water of a membrane electrode zygote becoming large by the above-mentioned configuration, humidifying [of ion exchange membrane] becoming easy since the diffusion (back diffusion of electrons) by the side of the negative electrode of the water generated with the positive electrode becomes easy to take place, and humidifying ion exchange membrane decreases, ion exchange membrane can fully be humidified only with generation water. For this reason, non-humidified actuation is attained, without making ion exchange membrane contain a noble-metals particle and a metallic-oxide particle.

[0018]

[Embodiment of the Invention] In the polymer electrolyte fuel cell equipped with the gas diffusion electrode which consists of the catalyst bed which formed the polymer electrolyte fuel cell of this invention in both sides of ion exchange membrane and said ion exchange membrane, and a diffusion layer, the periphery section of a catalyst bed is inside the periphery section of a diffusion layer, and thickness is 50 micrometers or less and ion exchange membrane is a polymer electrolyte fuel cell which has the reinforcement film of the shape of a frame which opened the aperture which is in agreement with the configuration of a catalyst bed in one [at least] field side.

[0019] Cross-section structural drawing of the polymer electrolyte fuel cell of one example of this invention is shown in drawing 1. As shown in drawing 1, the area of a catalyst bed 2 is smaller than the area of a diffusion layer 3, and it consists of ion exchange membrane 1 which has the reinforcement film 7 of the shape of a frame which prepared the gas diffusion electrode 4 which has the periphery section of a catalyst bed 2 inside the periphery section of a diffusion layer 3, and the window frame which is in agreement with the configuration of a catalyst bed 2

in one [at least] field side. 5 is a gasket and 7 is a separator.

[0020] By considering as such a configuration, since the reinforcement film 7 and the electrode catalyst bed 2 do not lap, the touch area of an electrode 4 and an ion exchange membrane 1 cannot be decreased, and the edge part of the electrode diffusion layer 3 can be located in the part in which the reinforcement film 7 exists. Therefore, even when thickness uses thin ion exchange membrane 50 micrometers or less, at the time of junction of the film and an electrode, breakage of the film at the time of the assembly of a cell and actuation is prevented, and it becomes possible to raise seal nature.

[0021] Furthermore, this invention is a polymer electrolyte fuel cell with which a positive electrode is supplied by oxidant gas and it supplies fuel gas to a negative electrode by no humidifying.

[0022] In a polymer electrolyte fuel cell, internal resistance becomes low and the outstanding cell property is shown, so that the ion exchange membrane of thin thickness is used.

Furthermore, by considering as such a configuration, the absolute magnitude of the water for humidifying ion exchange membrane becomes little, and the concentration gradient of water becomes large, the back diffusion of electrons of generation water becomes easy, and ion exchange membrane can fully be humidified only with generation water. Therefore, the non-humidified actuation of this solid-state polyelectrolyte mold fuel cell is attained.

[0023] In addition, although one example of this invention of drawing 1 showed what has the the same configuration of a catalyst bed and the the same window frame of a protective coat, what was mostly in agreement with the configuration of a catalyst bed shall be sufficient as a window frame, and it shall not lap with a catalyst bed. Moreover, even if a clearance exists in the window frame of a catalyst bed and a protective coat before assembly, it is possible for it to be also in agreement by binding each part article tight at the time of the assembly of a cell.

[0024] Moreover, although one example of this invention of drawing 1 showed what has the reinforcement film in one field side of ion exchange membrane, effectiveness with the same said of what has the reinforcement film in the both sides of ion exchange membrane as shown in drawing 2 is acquired.

[0025]

[Example] Hereafter, the example of this invention is explained, referring to a drawing.

[0026] (Example 1) The colloid dispersion liquid of the solid-state polyelectrolyte which mixed the alcoholic solution and organic solvent of a solid-state polyelectrolyte, and was agitated, and the carbon powder catalyst which made the platinum catalyst support were mixed, and it was made the shape of a paste, and applied on the carbon paper which gave a water-repellent finish with the fluororesin, and the forward negative electrode of the gas diffusion electrode which consists of a catalyst bed (50mmx50mm) and a diffusion layer (55mmx55mm) was created. The amount of platinum catalysts was made into 0.5 mg/cm² also with the forward negative electrode. Moreover, the amount of polyelectrolytes was made into 1.0 mg/cm² also with the forward negative electrode, and created the ion exchange membrane (80mmx80mm) equipped with the window frame which is 50mmx50mm the catalyst bed and whose configuration corresponded with the reinforcement film (80mmx80mm). The hotpress of said electrode and ion exchange membrane was carried out by 120-150 degrees C and 20 - 200 kgf/cm², and the membrane electrode zygote was produced. The ion exchange membrane used Nafion 112 (50 micrometers of thickness) by U.S. Du Pont. The reinforcement film was used as the sheet with adhesion material of a fluororesin with a thickness of 50 micrometers, which prepared the window frame of the same configuration as a catalyst bed, and was stuck on one side of ion exchange membrane. The cell A of the polymer electrolyte fuel cell equipped with the reinforcement film shown in drawing 1 using this zygote was produced.

[0027] (Example 1 of a comparison) The gas diffusion electrode which consists of 50mmx50mm the catalyst bed and diffusion layer of the same magnitude was created, except

having used the ion exchange membrane (80mmx80mm) which is not equipped with the reinforcement film, it presupposed that it is the same as an example 1, and the cell B without the reinforcement film shown in drawing 3 was produced.

[0028] (Example 2 of a comparison) The cell C made the same as an example 1 was produced except having used Nafion115 (130 micrometers of thickness) by U.S. Du Pont for the ion exchange membrane.

[0029] (Example 3 of a comparison) Except having prepared the window frame of the reinforcement film in 45mmx45mm so that the inner circumference of the reinforcement film might lap with the periphery section of a catalyst bed about 5mm, it presupposed that it is the same as an example 1, and the cell D shown in drawing 4 was produced.

[0030] (Example 2) It changed to Nafion112 (50 micrometers of thickness) by U.S. Du Pont which is the ion exchange membrane used in the example 1, and the cells E and F made the same as an example 1 were produced except having used Japan Gore-Tex GORE-SELECT (20 micrometers of thickness, 10 micrometers).

[0031] (Example 4 of a comparison) It changed to Nafion112 (50 micrometers of thickness) by U.S. Du Pont which is the ion exchange membrane used in the example 1 of a comparison, and the cells G and H made the same as the example 1 of a comparison were produced except having used Japan Gore-Tex GORE-SELECT (20 micrometers of thickness, 10 micrometers).

[0032] Above, hydrogen gas was supplied to the negative-electrode side of cell A-H of the example of this invention, and the example of a comparison, air was supplied to the positive-electrode side, and the spark test in non-humidified actuation and humidification actuation was performed. In non-humidified actuation, both hydrogen gas and air were supplied without no humidifying and warming, and did not perform warming of a cell, either. Moreover, in humidification actuation, hydrogen gas humidified and supplied air to 40 degrees C at 60 degrees C, and operating temperature was made into 50 degrees C.

[0033] The number of breakage of the ion exchange membrane in the cells A, E, and F of the example of this invention and the cells B, D, and H of the example of a comparison is shown in Table 1.

[0034]

[Table 1]

単電池	実 施 例			比 較 例			
	A	E	F	B	D	G	H
破損数/全数	0/10	0/10	0/10	2/10	0/10	6/10	10/10

[0035] Air and hydrogen were supplied, and when generation of heat was looked at by the no-load cell, it was judged that ion exchange membrane was damaged. Although breakage of ion exchange membrane was looked at by 2 in 10 cells in the cell B which does not use the reinforcement film when 50-micrometer ion exchange membrane was used, breakage of ion exchange membrane was not looked at by the cells A and D using the reinforcement film. Moreover, in the case where 20-micrometer ion exchange membrane is used, although

breakage of ion exchange membrane was looked at by 6 in 10 cells in the cell G which does not use the reinforcement film, breakage of ion exchange membrane was not looked at by the cell E using the reinforcement film. Although breakage of ion exchange membrane was looked at by all ten cells in the cell H which does not use the reinforcement film in the case where 10 more-micrometer ion exchange membrane is used, breakage of ion exchange membrane was not looked at by the cell F using the reinforcement film. When the reinforcement film was used, even if the thickness of ion exchange membrane was 50 micrometers or less, breakage did not arise from these results.

[0036] The current-voltage curve at the time of non-humidified actuation of the cells A, E, and F of the example of this invention and the cell C of the example of a comparison and humidification actuation is shown in drawing 5. Moreover, as compared with the time of humidification actuation, the cell voltage of non-humidified actuation fell with all cells. However, in dozens of mV and the cell C whose thickness is 130 micrometers to small one, the electrical-potential-difference difference was large and the sag in Cells A, E, and F fell remarkably with current density lower than especially 0.2 A/cm².

[0037] The current value at the time of non-humidified actuation of the cells A, E, and F of the example of this invention and the cell C of the example of a comparison and humidification actuation and the relation of internal resistance are shown in drawing 6. Moreover, although the internal resistance of non-humidified actuation increased with all cells as compared with the time of humidification actuation, as compared with Cells A, E, and F, the internal resistance of the large cell C of thickness increased remarkably.

[0038] In non-humidified actuation, ion exchange membrane is humidified only with generation water. There may be few moisture contents required in order to humidify ion exchange membrane so that thickness is small, if it is the same moisture content, as for the water content of ion exchange membrane, the direction of the film with thin thickness will become high, and resistance will become small. Moreover, if thickness is small, the concentration gradient of water will become large, it becomes easy to carry out the back diffusion of electrons of the water generated with the positive electrode, and ion exchange membrane becomes easy to be humidified. The thickness of the ion exchange membrane used for the cell A of this example is as thin as 50 micrometers. Therefore, it can be said that Cell A showed the polarization property which internal resistance becomes [the water content of ion exchange membrane] comparatively high low also by non-humidified actuation, and does not have humidification actuation and inferiority. With the cells E and F using ion exchange membrane with still thinner thickness, such effectiveness becomes large, non-humidified actuation becomes easy, and it is thought that the property still higher than Cell A was shown. since Cell C, on the other hand, has the thickness of ion exchange membrane as thick as 130 micrometers -- generation water -- humidification of ion exchange membrane -- inadequate -- internal resistance -- high -- becoming -- especially -- generation -- it appears notably in a low current consistency region with little amount of water, and the property of non-humidified actuation is considered to have fallen as compared with humidification actuation.

[0039] The current-voltage curve in non-humidified actuation of the cell A of the example of this invention and the cells B and D of the example of a comparison is shown in drawing 7. The open-circuit voltage of Cells A, B, and D is 1015mV, 900mV, and 1010mV, respectively, and Cells A and D showed the value higher than Cell B. Moreover, Cells A and B are the almost same polarization properties, and showed the property superior to Cell D.

[0040] Since Cells A and D showed open-circuit voltage higher than Cell B, it can be said that the crossover of the reactant gas from the periphery of an electrode was controlled with the reinforcement film. Moreover, in Cell D, since the reinforcement film has lapped with the electrode catalyst bed, the area of an electrode and ion exchange membrane decreases. However, Cell A can be said to have shown the polarization property which ion exchange

membrane was reinforced and was superior to Cell D, without an electrode surface product decreasing, since the reinforcement film does not lap with an electrode catalyst bed.

[0041] In addition, although the Nafion film by U.S. Du Pont and Japan Gore-Tex GORE-SELECT were used for the ion exchange membrane in this example, if it is the ion exchange membrane which shows cation conductivity, it will not be limited to this.

[0042] Moreover, although the fluoro-resin sheet was used for the reinforcement film, the same effectiveness will be acquired if it has thermal resistance and acid resistance. Although the sheet which has adhesion material was used in this example, the same effectiveness is acquired also by uniting with ion exchange membrane the sheet which does not have adhesion material by thermal melting arrival etc. Although the reinforcement film with a thickness of 50 micrometers was furthermore used by this example, it is necessary to restrict the thickness of the reinforcement film with the thickness in an example, but to optimize it with the thickness of ion exchange membrane.

[0043] Although the periphery section of the reinforcement film and the periphery section of ion exchange membrane are furthermore in agreement by this example, the same effectiveness will be acquired if the periphery section of the reinforcement film is outside the periphery section of a gaseous diffusion layer.

[0044]

[Effect of the Invention] As mentioned above, without decreasing the reaction area of an electrode, when the ion exchange membrane of a thin film is used, according to this invention, at the time of membrane electrode junction, breakage of ion exchange membrane can be prevented at the time of cell assembly and actuation, and the polymer electrolyte fuel cell which has the outstanding seal nature can be realized.

[0045] Moreover, it becomes possible to perform non-humidified actuation of a polymer electrolyte fuel cell, without the hydrogen utilization factor by making hydrogen and oxygen react in ion exchange membrane decreasing.

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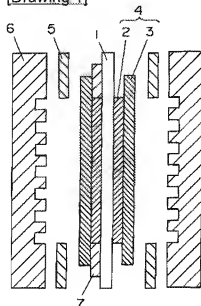
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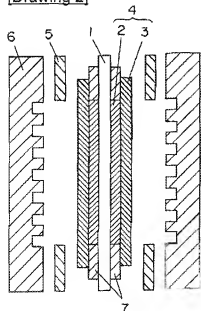
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DRAWINGS

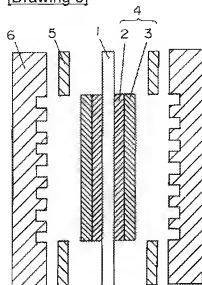
[Drawing 1]



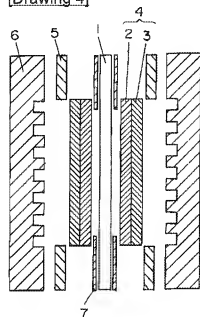
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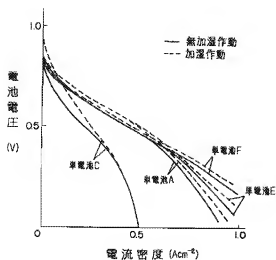
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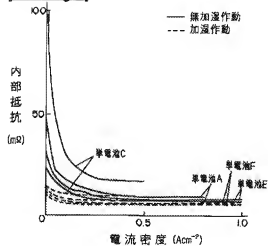
[Drawing 4]



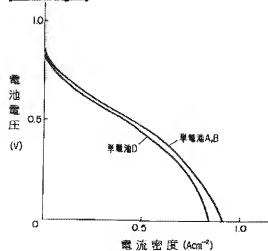
[Drawing 5]



[Drawing 6]



[Drawing 7]



[Translation done.]